REMARKS

Claims 1, 2, 4-9, 11 and 13-18 remain pending in this application. Claims 3, 10 and 12 have been canceled without prejudice or disclaimer.

Claim Amendments

In order to more clearly define the present invention, Applicants have amended claim 1 as follows.

The reactive organometal compound used in step (1) has been limited to at least one organometal compound selected from the group consisting of an organometal compound represented by formula (1) and an organometal compound represented by formula (2), each of which was recited in claim 3. Also, in this amendment, the organometal compound of formula (1) and the organometal compound of formula (2) have been limited so that the hydrocarbon groups (i.e., R^3 and R^4) constituting the "OR" groups (i.e., OR^3 and OR^4) of the organometal compound of formula (1) and the hydrocarbon groups (i.e., R^9 and R^{10}) constituting the "OR" groups (i.e., OR^9 and OR^{10}) of the organometal compound of formula (2) are the same.

By incorporating in claim 1 the feature that R^3, R^4, R^9 and R^{10} are the same, a symmetric carbonic ester (i.e., a carbonic ester represented by the formula: RO(CO)OR) can be obtained, as explained below.

The carbonic ester is produced by the reaction between the reactive organometal compound and carbon dioxide performed in step (1). In this connection, the Examiner's attention is drawn to the following description of the present specification:

> "In the method of the present invention, an asymmetric carbonic ester can be produced as follows. Explanations are given below, taking as an example the case where the

> reactive organometal compound has at least one type of alkoxy group. When the reactive organometal compound used in step (1) has two different types of alkoxy groups, an asymmetric carbonic ester can be produced without use of alcohols (as a second alcohol and a third alcohol) in steps (1) and (2)." (emphasis added) (see page 89, lines 9-18 of the present specification)

The above-cited passage of the present specification states that when the reactive organometal compound used in step (1) has two different types of "OR" groups, the reaction between the reactive organometal compound and carbon dioxide produces an asymmetric carbonic ester. In other words, when the reactive organometal compound has only one type of "OR" group, the reaction between the reactive organometal compound and carbon dioxide produces a symmetric carbonic ester (i.e., a carbonic ester represented by the formula: RO(CO)OR). More specifically, when R^3 and R^4 in the organometal compound of formula (1) are the same, the reaction between the organometal compound of formula (1) and carbon dioxide produces a symmetric carbonic ester represented by the formula: $R^3O(CO)OR^3$; and when R^9 and R^{10} in the organometal compound of formula (2) are the same, the reaction between the organometal compound of formula (2) and carbon dioxide produces a symmetric carbonic ester represented by the formula: $R^9O(CO)OR^9$.

When the organometal compound of formula (1) and the organometal compound of formula (2) are used individually, R^3 in the organometal compound of formula (1) and R^9 in the organometal compound of formula (2) may be different. However, when the organometal compound of formula (1) and the organometal compound of formula (2) are used simultaneously, the feature that R^3 and R^9 are the same (i.e., the feature that

R^3, R^4, R^9 and R^{10} are the same) is required. On this point, an explanation is given below.

As explained above, the reaction between the organometal compound of formula (1) (wherein R^3 and R^4 are the same) and carbon dioxide produces a symmetric carbonic ester represented by the formula: $R^3O(CO)OR^3$, and the reaction between the organometal compound of formula (2) (wherein R^9 and R^{10} are the same) and carbon dioxide produces a symmetric carbonic ester represented by the formula: $R^9O(CO)OR^9$. If R^3 and R^9 are different, an asymmetric carbonic ester $R^3O(CO)OR^9$ is also produced by the transesterification reaction between the symmetric carbonic esters $R^3O(CO)OR^3$ and $R^9O(CO)OR^9$. In order to prevent the production of such asymmetric carbonic ester, the incorporation of the feature that R^3, R^4, R^9 and R^{10} are the same is necessary.

The fact that a symmetric carbonic ester is produced by the reaction between carbon dioxide and the reactive organometal compound recited in amended claim 1 is substantiated by the working examples of the present application. Specifically, for example, in the case of Example 1 of the present application, dibutyltin di(2-ethylhexyloxide) (which is a compound of formula (1) wherein R^3 and R^4 are the same) and 1,1,3,3-tetrabutyl-1,3-di(2-ethylhexyloxy)distannoxane (which is a compound of formula (2) wherein R^9 and R^{10} are the same) are simultaneously used as reactive organometal compounds for the reaction with carbon dioxide to produce di(2-ethylhexyl) carbonate, which is a symmetric carbonic ester (see page 121, lines 16-18 and page 122, lines 18-19 of the present specification).

From the above, it is apparent that a symmetric carbonic ester is produced by the method of amended claim 1.

In accordance with the amendment to claim 1, claim 3 has been canceled, and claims 4-6 have been amended. Further, claims 10 and 12 have been canceled. No new matter has been introduced by these amendments.

Priority and IDS citations acknowledgement

Applicants note, with appreciation, that the examiner has acknowledged the claim for foreign priority, and acknowledged receipt and consideration of documents cited in the Information Disclosure Statement.

Abstract:

On page 2, paragraph 2 of the Office action, the Office requires correction of the abstract that is alleged to contain more than 150 words. This is not correct. The abstract appearing as a separate page numbered 152 is about 144 words and clearly in compliance with 37 C.F.R. § 1.72(b). Accordingly, this objection should be withdrawn.

Restriction - election

Applicants acknowledge election of the subject matter of Group I for examination. Pursuant to the election of species, applicants elected dibutyltin bis(3-methylbutoxide). All pending claims (1, 2, 4-9, 11 and 13-18) are readable on the elected species.

Although applicants have amended the claims to be commensurate in scope with the elected invention, applicants reserve the right to file a continuing application directed to the non-elected subject matter. Accordingly, the objection to claims 1-18 in paragraph 11 of the Office action should be withdrawn.

Rejection - § 112, first paragraph

Claims 1-18 have been rejected under 35 U.S.C. § 112, first paragraph, because the specification is allegedly nonenabling for processes of making carbonic ester

compounds or organometal compounds without limitation of the structural formula. While applicants disagree with this position, in order to expedite prosecution of this application, the claims have been amended to be limited to the subject matter that the examiner has acknowledged is enabled by the present specification. Accordingly, this rejection should be withdrawn.

<u>Rejection - § 112, second paragraph</u>

Claims 1-18 have been rejected under 35 U.S.C. § 112, second paragraph, as being indefinite as step (3) of claim 1 does not state how the final product carbonic ester is obtained from the second organometal compound mixture. Clarification is required.

An isolated carbonic ester as the final product is <u>not</u> obtained by the operation performed in step (3). The carbonic ester is formed in step (1) and, after the operation of step (2), is present in the form of a mixture referred to as "first portion".

The method for separating the carbonic ester from the first portion is not described in claim 1, because the separation of the carbonic ester from the first portion can be performed easily by any conventional method. For example, a relevant portion of the present specification is reproduced below:

> " The separation of the carbonic ester from the first portion (containing the carbonic ester and the unregenerable unreactive compound) obtained in step (2) can be performed easily by any of the conventional methods, such as adsorption, distillation, filtration and membrane separation." (see page 96, lines 15-20 of the present specification)

Consequently, although a carbonic ester is produced by the claimed method, as recited in the preamble, it is not produced in separated form. Accordingly, this rejection should be withdrawn as all pending claims are clear.

State of the art and the present invention

Before specifically addressing the Examiner's rejections under 35 U.S.C. 102(b) or 103(a), it is believed that the following background information should be considered in order to shed proper light on the development of the present invention and the advantageous features thereof.

As described in the present specification under "Prior Art", there are a number of methods for producing a carbonic ester. In these methods, carbon dioxide is subjected to an equilibrium reaction with an alcohol in the presence of an organometal compound as a catalyst, thereby producing a carbonic ester and water. As explained at page 6, lines 9-21 of the present specification, this equilibrium reaction is represented by the following formula (3):

$$CO_2 + 2ROH \underset{}{\overset{\text{organometal compound}}{\rightleftarrows}} RO(CO)OR + H_2O \quad (3)$$

(R represents an unsaturated or saturated hydrocarbon group)

The conventional methods using the reaction of formula (3) above have the following disadvantage. The water formed by the reaction decomposes the catalyst, thereby depriving the catalyst of its catalytic activity. When the catalyst has lost its catalytic activity, there is no way to easily and effectively regenerate and reuse the catalyst. For preventing the decomposition of the catalyst so as to make it possible to reuse the catalyst, it is necessary to remove water from the reaction system by the use of a dehydrating agent. Thus, the above-mentioned conventional methods are

disadvantageous in that the use of a dehydrating agent is necessary for making it possible to reuse the catalyst.

In this situation, the present inventors have made extensive and intensive studies for solving the above-mentioned problem. As a result, it has unexpectedly been found that the problem can be solved by a method as described in amended claim 1 of the present application. This method is a method for producing a carbonic ester, which comprises the steps of:

(1) performing a reaction between a first organometal compound mixture and carbon dioxide, wherein the first organometal compound mixture comprises a mixture of a reactive organometal compound and an unregenerable unreactive compound derived from the reactive organometal compound, to thereby obtain a reaction mixture containing a carbonic ester formed by the reaction, the unregenerable unreactive compound, and a regenerable metamorphic organometal compound derived from the reactive organometal compound,

(2) separating the reaction mixture into a first portion containing the carbonic ester and the unregenerable unreactive compound, and a second portion containing the regenerable metamorphic organometal compound, and

(3) reacting the second portion of the reaction mixture with an alcohol to form a second organometal compound mixture and water and removing the water from the second organometal compound mixture, wherein the second organometal compound mixture comprises a mixture of a reactive organometal compound and an unregenerable unreactive compound derived from the reactive organometal compound.

The reaction performed in step (1) is represented by the following formula (4) (see page 33 of the present specification):

$$\text{organometal compound} + CO_2 \rightleftarrows \text{CO_2 adduct of organometal compound} \xrightleftharpoons{\text{thermal decomposition}} \text{carbonic ester + other thermal decomposition products} \qquad (4)$$

It should be noted that the reactive organometal compound recited in step (1) of the method of the present invention is not used as a catalyst, but as a reagent to be reacted with carbon dioxide, differing from the case of the conventional methods using the reaction of formula (3) above, in which an organometal compound is used as a catalyst.

In the method of the present invention, a reactive organometal compound as recited in step (1) is regenerated in step (3) and, hence, can be recycled to step (1). Therefore, the method of the present invention is advantageous in that a reactive organometal compound can be reused without the need for the use of a dehydrating agent. This is a great advantage of the present invention over the conventional methods.

The method of the present invention is also advantageous in that an unregenerable unreactive compound formed during the reaction can be removed from the reaction system and, hence, the accumulation of the unregenerable unreactive compound in the reaction system can be prevented. This advantage is described in the

present specification (see page 35, line 24 to page 38, line 11 of the present specification).

References cited as prior art

(1) Itakura et al. (JP 11035521) (hereinafter, frequently referred to as "JP '521");

(2) Ko et al. (JP 07033715) (hereinafter, frequently referred to as "JP '715");

(3) Yamazaki et al. (JP 54003012) (hereinafter, frequently referred to as "JP '012"); and

(4) U.S. Patent Application No. 10/495,451 (hereinafter, frequently referred to as "Miyake '451").

JP '521, JP '715 and JP '012 are Japanese language patent documents. For the Examiner's convenience, Applicants submit full or partial English translations of these Japanese language patent documents. Specifically, with respect to JP '521 and JP '715, Applicants submit full English translations thereof as Exhibits 1 and 2, respectively, which are machine translations provided by the Japan Patent Office. With respect to JP '012, no such English translation has been provided by the Japan Patent Office and, hence, Applicants submit a sworn partial English translation of JP '012 as Exhibit 3.

Arguments addressing JP '521, JP '715 and JP '012 are made with reference to Exhibits 1 to 3.

Rejections - §§ 102/103 Itakura et al.

Claims 1-18 have been rejected as being anticipated under 35 U.S.C. § 102(b) or as obvious under 35 U.S.C. § 103(a) over Itakura et al. (JP '521). These rejections are traversed for the following reasons.

JP '521 discloses a method for producing a carbonic ester. The abstract of JP '521 states that, in the method of JP '521, a carbonic ester is produced by reacting a carboxylic acid orthoester with carbon dioxide in the presence of a metal alkoxide and a halide as a catalyst (see the abstract of JP '521, an English translation of which is shown in Exhibit 1).

However, the method of JP '521 (JP 11035521) actually resides in that an alcohol is reacted with carbon dioxide to produce a carbonic ester and water, wherein the water is removed from the reaction system by the use of a carboxylic acid orthoester as a dehydrating agent. This is apparent from the following description of the present specification:

> " Another method for producing a carbonic ester uses a carboxylic acid orthoester as an organic dehydrating agent (see <u>Unexamined Japanese Patent Application Laid-Open Specification No. Hei 11-35521</u>). (In this patent document, there are descriptions reading: "a carboxylic acid orthoester is reacted with carbon dioxide" and "an acetal is reacted with carbon dioxide". However, as a result of recent studies in the art, it is generally presumed that the actual reaction route is as follows. "An alcohol and carbon dioxide are reacted with each other to obtain a carbonic ester and water. The water is reacted with a carboxylic acid orthoester.") This method has problems in that a carboxylic acid orthoester (which is an expensive compound) is used as a dehydrating agent, and methyl acetate is by-produced (see "Kagaku Sochi (Chemical Equipment)", Vol. 41, No.2, 52-54 (1999)). Thus, this method is as defective as the above-mentioned methods." (emphasis added) (see page 8, lines 8-25 of the present specification)

Therefore, the method of JP '521 is one example of the conventional methods using the reaction of formula (3) above. JP '521 has no teaching or suggestion about step (1), (2) or (3) of the method of the present invention. Especially, it should be noted

that the method of JP ’521 uses an organometal compound (metal alkoxide) as a catalyst, differing from the method of the present invention in which a reactive organometal compound is used as a reagent to be reacted with carbon dioxide.

JP ’521 has the same problem as encountered in the conventional methods using the reaction of formula (3) above. JP ’521 does not teach or suggest how to solve this problem. The problem has for the first time been solved by the present invention.

Thus, JP ’521 has no teaching that describes the steps as claimed, and there is no reason or suggestion to modify the process of JP ‘521 to obtain the present invention and the effects achieved thereby. Accordingly, these rejections should be withdrawn.

Rejections - §§ 102/103 Ko et al.

Claims 1-18 have been rejected as being anticipated under 35 U.S.C. § 102(b) or as obvious under 35 U.S.C. § 103(a) over Ko et al. (JP ‘715). These rejections are traversed for the following reasons.

JP ’715 discloses a method for producing a carbonic ester, in which an alcohol is reacted with carbon dioxide in the presence of an organometal compound as a catalyst, wherein water produced by the reaction is removed from the reaction system by the use of a dehydrating agent, such as an orthoformic triester or molecular sieves (see the abstract, claim 1 and paragraph [0010] of JP ’715, an English translation of which is shown in Exhibit 2).

Therefore, the method of JP ’715 is one example of the conventional methods using the reaction of formula (3) above. JP ’715 has no teaching or suggestion about step (1), (2) or (3) of the method of the present invention. Especially, it should be noted that the method of JP ’715 uses an organometal compound as a catalyst, differing from

the method of the present invention in which a reactive organometal compound is used as a reagent to be reacted with carbon dioxide.

JP '715 has the same problem as encountered in the conventional methods using the reaction of formula (3) above. JP '715 does not teach or suggest how to solve this problem. The problem has for the first time been solved by the present invention.

Thus, JP '715 has no teaching that describes the steps as claimed, and there is no reason or suggestion to modify the process of JP '715 to obtain the present invention and the effects achieved thereby. Accordingly, these rejections should be withdrawn.

Rejections - §§ 102/103 Yamazaki et al.

Claims 1-18 have been rejected as being anticipated under 35 U.S.C. § 102(b) or as obvious under 35 U.S.C. § 103(a) over Yamazaki et al. (JP '012). These rejections are traversed for the following reasons.

JP '012 discloses a method for producing a carbonic ester, which comprises reacting a hydroxy compound with carbon dioxide in the presence of an organometal compound (specifically a tin alkoxide or a tetraalkoxytitanium) as a catalyst (see claim 1 of JP '012, an English translation of which is shown in Exhibit 3). As the hydroxy compound, an alcohol or phenolic compound can be used (see a relevant portion of JP '012, an English translation of which is shown in Exhibit 3). Therefore, the reaction performed in the method of JP '012 is the same or substantially the same as the reaction of formula (3) above.

Accordingly, the method of JP '012 is considered to be one example of the conventional methods using the reaction of formula (3) above. JP '012 has no teaching or suggestion about step (1), (2) or (3) of the method of the present invention.

Especially, it should be noted that the method of JP '012 uses an organometal compound (specifically a tin alkoxide or a tetraalkoxytitanium) as a catalyst, differing from the method of the present invention in which a reactive organometal compound is used as a reagent to be reacted with carbon dioxide.

JP '012 has the same problem as encountered in the conventional methods using the reaction of formula (3) above. JP '012 does not teach or suggest how to solve this problem. The problem has for the first time been solved by the present invention.

Thus, JP '012 has no teaching that describes the steps as claimed, and there is no reason or suggestion to modify the process of JP '012 to obtain the present invention and the effects achieved thereby. Accordingly, these rejections should be withdrawn.

Rejection - provisional double patenting

Claims 1-18 are provisionally rejected under the judicially-created doctrine of obviousness-type double patenting as being unpatentable over claim 1 of Miyake et al. (copending Application No. 10/495,451). In order to expedite prosecution, applicants are filing a terminal disclaimer to avoid this rejection. The filing of a terminal disclaimer is not an admission of the propriety of the rejection. *Quad Environmental Technologies Corp. v. Union Sanitary District*, 946 F.2d 870 (Fed. Cir. 1991). Accordingly, this rejection should be withdrawn.

Prompt and favorable reconsideration of this application is respectfully requested.

Please grant any extensions of time required to enter this response and charge any additional required fees to our deposit account 06-0916.

Respectfully submitted,

FINNEGAN, HENDERSON, FARABOW,
GARRETT & DUNNER, L.L.P.

Dated: March 28, 2008

By: Charles E. Van Horn
Charles E. Van Horn
Reg. No. 40,266

Attachments: Exhibits 1-3
Terminal Disclaimer

Exhibit 1

PATENT ABSTRACTS OF JAPAN

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(71)Applicant : AGENCY OF IND SCIENCE & TECHNOL
(72)Inventor : SAKAKURA TOSHIYASU
SAKO TAKESHI

(54) PRODUCTION OF CARBONATE

(57)Abstract:
PROBLEM TO BE SOLVED: To produce carbonate in high yield by reacting carbon dioxide with carboxylic acid orthoester in the presence of metal alkoxide and halide.
SOLUTION: This objective compound is obtained by reacting carbon dioxide with carboxylic acid orthoester (e.g. methyl orthoformate) shown by the formula R1C(OR2)3 (R1 is H, etc.; and R2 is an alkyl, etc.), in the presence of a metal alkoxide [e.g. Sn(OMe)4(Me is a methyl), etc.], shown by the formula (R3)nM (OR4)4-n (R3 and R4 are each an alkyl, etc.; (n) is 0 to 3; and M is a metal) and halide (e.g. quaternary phosphonium salt or quaternary ammonium salt) preferably at room temperatures to 20° C under the pressure of 1 to 500 atm. for 1 to 100 hrs.

CLAIMS

[Claim(s)]
[Claim 1]A manufacturing method of carbonic ester making carbon dioxide and carboxylic acid ortho ester react under existence of a metal alkoxide and a halogenide.
[Claim 2]A manufacturing method of the carbonic ester according to claim 1 which is an alkoxide of metal in which a metal alkoxide is chosen from tin, titanium, or a zirconium.
[Claim 3]A manufacturing method of the carbonic ester according to claim 1 or 2 whose halogenide is the fourth class phosphonium salt, alkali metal salt, or quarternary ammonium salt.
[Claim 4]A manufacturing method of carbonic ester making carboxylic acid ortho ester react to carbon dioxide under existence of a halogenide chosen from the fourth class phosphonium salt or alkali metal salt.
[Claim 5]A manufacturing method of the carbonic ester according to claim 3 or 4 which uses alkali metal salt with a crown ether compound.
[Claim 6]A manufacturing method of the carbonic ester according to claim 1, 2, 3, 4, or 5 making carboxylic acid ortho ester react to carbon dioxide under existence of Lewis acid.

[Translation done.]

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention]This invention relates to the method of manufacturing carbonic ester from carbon dioxide and carboxylic acid ortho ester.
[0002]
[Description of the Prior Art]Carbonic ester is a compound useful as the diesel fuel additives for decreasing the gasoline additive for raw materials, such as polycarbonate manufacture, and the improvement in the octane number, and the particle in exhaust gas, an alkylating agent, a carbonylation agent, a solvent, etc. Although the method of making phosgene into a carbonylation agent and making it react to alcohol first as a manufacturing method of conventional carbonic ester is raised, In this method, in order that toxicity might use extremely the phosgene which also has corrosiveness strongly, cautions are required for handling, such as that transportation, storage, etc., and cost great for safety reservation of the control of maintenance and waste treatment of a manufacturing facility, and authorized personnel etc. had started. Although the oxidative carbonylating method make carbon monoxide into a carbonylation agent and make it react to alcohol and oxygen is also known, In order to use carbon monoxide also in this method, cautions were required because of safety reservation of authorized personnel etc., and carbon monoxide oxidized and there was a fault from which the side reaction of generating carbon dioxide occurs. For this reason, development of the method of manufacturing carbonic ester more safely and at a low price is demanded, The method of making carbon dioxide into a carbonylation agent and making it react to alcohol was proposed (Applied Catalysis, 1996, and 142 L 1 page;Collect. Czech. Chem. Commun. a magazine, 1995, 60 volumes, 687 pages, etc.). However, 2 or about 3, and catalytic activity had the very low turnover number, the water to generate decomposed the catalyst and any method had a problem of checking a reaction. the method of manufacturing carbonic ester from the reaction of carbon dioxide and carboxylic acid ortho ester is also proposed -- **** (JP,7-244010,A) -- there was a technical problem in industrial operation -- catalytic activity is low.
[0003]
[Problem(s) to be Solved by the Invention]Therefore, an object of this invention is to provide the method that carbonic ester can be industrially manufactured with high yield by making into a carbonylation agent carbon dioxide which there are not toxicity and corrosiveness and is obtained very at a low price.
[0004]
[Means for Solving the Problem]If this invention person uses a metal alkoxide and a halogenide as a catalyst in a reaction of carbon dioxide and carboxylic acid ortho ester as a result of repeating research wholeheartedly, in order to solve a problem of the above-mentioned conventional method, It finds out that carbonic ester is obtained at high catalyst efficiency, and came to make this invention based on this knowledge. This invention Namely, under existence of (1) metal alkoxide and a halogenide, A manufacturing method of carbonic ester making carbon dioxide and carboxylic acid ortho ester react, (2) A manufacturing method of carbonic ester given in (1) paragraph a given metal alkoxide is an alkoxide of metal chosen from tin, titanium, or a zirconium, (3) (1) or a manufacturing method of carbonic ester given in (2) paragraphs whose halogenide is the fourth class phosphonium

salt, alkali metal salt, or quarternary ammonium salt, (4) Under existence of a halogenide chosen from the fourth class phosphonium salt or alkali metal salt, A manufacturing method of carbonic ester making carboxylic acid ortho ester react to carbon dioxide, (5) (3) which uses alkali metal salt with a crown ether compound, or a manufacturing method of carbonic ester given in (4) paragraphs, And a manufacturing method of carbonic ester (1), (2), (3), (4), or given in (5) paragraphs making carboxylic acid ortho ester react to carbon dioxide is provided under existence of (6) Lewis acid.
[0005]
[Embodiment of the Invention]In this invention, carboxylic acid ortho ester is made to react to carbon dioxide, and carbonic ester is manufactured. The carboxylic acid ortho ester which can be used by this invention is expressed with following general formula (I).
[0006]General formula (I)
$R^1C(OR^2)_3$ (as for R^1, hydrogen or an alkyl group, and R^2 express an alkyl group, an alkenyl group, or an aryl group among a formula.)
[0007]Among general formula (I), the alkyl group expressed with R^1 and R^2 is a low-grade alkyl group preferably, and are the carbon numbers 1-4 still more preferably. Specifically, methyl, ethyl, n-propyl, n-butyl, etc. are mentioned. the alkenyl group expressed with R^2 -- desirable -- the carbon numbers 2-6 -- it is 2-3 still more preferably, for example, allyl, vinyl, etc. are mentioned. the aryl group expressed with R^2 -- desirable -- the carbon numbers 6-14 -- it is 6-10 still more preferably, for example, phenyl, tolyl, anisyl, naphthyl, etc. are mentioned. More specifically as such carboxylic acid ortho ester, alt.methyl formate, ethyl orthoformate, alt.formic acid allyl, alt.formic acid phenyl, alt.methyl acetate, alt.ethyl acetate, alt.allyl acetate, alt.phenyl acetate, etc. are mentioned, for example.
[0008]The reaction of this invention can be performed under existence of a metal alkoxide and a halogenide. The metal alkoxide used here is expressed with following general formula (II).
[0009]General formula (II)
$(R^3)_nM(OR^4)_{4-n}$ (R^3 and R^4 express an alkyl group, an alkenyl group, or an aryl group among a formula, and n expresses the integer of 0-3.) M expresses a metal atom.
[0010]The alkyl group expressed with R^3 and R^4 is a low-grade alkyl group preferably, and are the carbon numbers 1-4 still more preferably. Specifically, methyl, ethyl, n-butyl, isopropyl, hexyl, cyclohexyl, etc. are mentioned. The alkenyl groups expressed with R^3 and R^4 may be the carbon numbers 2-10 preferably, and a chain and annular any may be sufficient as them. Specifically, cyclopentadienyl, pentamethylcyclopentadienyl, indenyl, vinyl, allyl, etc. are mentioned. The aryl groups expressed with R^3 and R^4 are the carbon numbers 6-14 preferably, for example, phenyl, tolyl, anisyl, naphthyl, etc. are mentioned. As a metal atom expressed with M, tin, titanium, and a zirconium are preferred. It is made to generate by making a corresponding metaled halogenide and alkaline metal alkoxides, such as sodium methoxide and magnesium methoxide, react in a system, and these metal alkoxides can also be used. Although the example of a metal alkoxide is given to below, this invention is not limited to this.
[0011]Sn. $(OMe)_4Bu_2Sn.$ $(OMe)_2Bu_2Sn.$ $(OEt)_2Bu_2Sn.$ $(OBu)_2Bu_3Sn.$ (OMe) Ti. $(OMe)_4Ti.$ $(O\text{-}i\text{-}Pr)_4Ti.$ $(OBu)_4Cp_2Ti.$ $(OMe)_2Cp_2Ti(OPh)_2Zr(OMe)_4Zr(O\text{-}i\text{-}Pr)_4Zr(OBu)_4Cp_2Zr(OMe)_2Cp_2Zr(OPh)_2$
(Me: -- methyl and Et:ethyl.) i-Pr: Isopropyl, Bu:n-butyl, Cp:cyclopentadienyl, Ph:phenyl
[0012]As a halogenide used above, the fourth class phosphonium salt, quarternary ammonium salt, alkali metal salt, or bis(triphenyl phosphoranyliden)ammonium salt is mentioned. As the fourth class phosphonium salt, tetra alkyl phosphonium salt, tetra aryl phosphonium salt, etc. can be used, and a tetrabutylphosphonium salt, tetraoctyl phosphonium salt, etc. are specifically mentioned. As quarternary ammonium salt, tetra-alkyl ammonium salt, tetra aryl ammonium salt, etc. can be used, and a tetrabutylammonium salt, tetraoctylammonium salt, the Millis Chill trimethylammonium salt, etc. are specifically mentioned. As alkali metal salt, potassium salt, sodium salt, etc. are mentioned, for example. As halogen of such a halogenide, although chlorine, iodine, bromine, etc. are raised, iodine is preferred. Since solubility is low when using alkali metal salt as a halogenide, it is preferred to make a crown ether compound, cryptand, etc. live together as a host compound, and it is still more preferred

to make a crown ether compound live together. As a crown ether compound, the 9-crown 3, the 12-crown 4, the 15-crown 5, the 18-crown 6, etc. are mentioned, for example, and it may have a substituent. Such a crown ether compound and a complex compound with lithium, sodium, potassium, etc. can also be used. About cryptand, [2.2.1]-cryptand, [2.2.2]-cryptand, etc. are specifically mentioned and a complex with these metal ions can also be used.
[0013]The reaction of carbon dioxide and carboxylic acid ortho ester can be performed under existence of the fourth class phosphonium salt or alkali metal halides. What was raised with the aforementioned halogenide is raised as the fourth class phosphonium salt in this case, and an example of alkali metal halides.
[0014]the reaction of the carbon dioxide in this invention, and carboxylic acid ortho ester -- usually -- room temperature - it is 80-150 ** preferably, and 200 ** is performed for 1 to 100 hours. 1-500 atmospheres of inside of the system of reaction react by being filled up with carbon dioxide so that it may become 9.5-300 atmospheres preferably. Although a solvent in particular is not needed, the solvent which does not check reactions, such as hexane, benzene, and methanol, can also be used. In this invention, the amount of the above-mentioned metal alkoxide, a halogenide, the fourth class phosphonium salt, or the alkali metal halides used is what is called a catalyst amount, and is usually 1/100,000 - 1(mol)/10 to carboxylic acid ortho ester. The generated carbonic ester can isolate in accordance with conventional methods, such as distillation.
[0015]Even when the reaction of the carbon dioxide in this invention and carboxylic acid ortho ester has no Lewes acid catalyst, it goes on, but reacting under existence of Lewis acid is also preferred. As Lewis acid at this time, the following compounds are raised, for example.
BF_3 and $OEt_2La(OSO_2CF_3)\ _3Ph_3C^+B(C_6F_5)\ _4{}^-$ (Et: ethyl, Ph:phenyl)
[0016]
[Example]Next, this invention is explained still in detail based on an example.
To the autoclave made from SUS of 20 ml of inner capacity with example 1 agitating equipment. As a metal alkoxide, dibutyl-tin-dimethoxide 0.85mmol, Tetrabutylammonium iodide 0.81mmol and alt.methyl-acetate 49.5mmol were taught as a halogenide, it was filled up with the liquefied carbon dioxide from the carbon dioxide cylinder, and internal pressure was adjusted to 65kg[/cm] ^{2}. Then, it heated at 150 **, stirring the inside of autoclave, and was made to react for 24 hours. The pressure in the autoclave at the time of a pyrogenetic reaction was a maximum of 250kg[/cm] ^{2}. When the carbon dioxide which remains was emitted after cooling and gas chromatography analyzed reaction mixture, generation of 11.22% of dimethyl carbonate was checked with the yield to alt.methyl acetate.
[0017]When reacted completely like Example 1 except not using a comparative example 1 metal alkoxide, the yield of dimethyl carbonate was 5.27% and there was far compared with Example 1.
[little]
[0018]When reacted like Example 1 using the ortho ester, the metal alkoxide, halogenide, and solvent which are shown in Examples 2-8, the comparative example 2 - the 4 following table 1, dimethyl carbonate generated with the yield (as opposed to the charge of alt.methyl acetate) shown in Table 1.
[0019]
[Table 1]

表 1

No.	温度 (℃)	時間 (h)	オルトエステル 化合物	オルトエステル (mmol)	触媒 化合物	触媒 (mmol)	触媒 化合物	触媒 (mmol)	溶媒 化合物	溶媒 (mmol)	炭酸ジメチル 収率*1 (%)
実施例1	150	24	$CH_3C(OCH_3)_3$	49.5	$Bu_2Sn(OMe)_2$	0.85	Bu_4NI	0.81	なし		11.22
比較例1	160	24	$CH_3C(OCH_3)_3$	49.5	なし		Bu_4NI	0.81	なし		5.27
比較例2	150	24	$CH_3C(OCH_3)_3$	49.5	$Bu_2Sn(OMe)_2$	0.85	なし		なし		0.84
実施例2	150	24	$CH_3C(OCH_3)_3$	49.5	$Bu_2Sn(OMe)_2$	0.85	Bu_4NI	0.81	メタノール	41	19.22
実施例3	150	24	$CH_3C(OCH_3)_3$	49.5	$Bu_3Sn(OMe)$	0.86	Bu_4NI	0.81	メタノール	41	5.69
実施例4	150	24	$HC(OCH_3)_3$	50.3	$Bu_2Sn(OMe)_2$	0.85	Bu_4NI	0.81	メタノール	41	1.21
比較例3	150	24	$CH_3C(OCH_3)_3$	49.5	Bu_2SnI_2	0.85	なし		なし		0.02
実施例5	150	24	$CH_3C(OCH_3)_3$	49.5	$Bu_2Sn(OMe)_2$	0.87	Bu_4PI	0.85	なし		15.36
実施例6	150	24	$CH_3C(OCH_3)_3$	49.5	なし		Bu_4PI	0.85	なし		6.35
実施例7	150	24	$CH_3C(OCH_3)_3$	49.5	$Bu_2Sn(OMe)_2$	0.87	KI クラウンエーテル*2	0.88 0.89	なし		2.42
実施例8	150	24	$CH_3C(OCH_3)_3$	49.5	$Cp_2Ti(OMe)_2$	0.86	$Mg(OMe)_2$ Bu_4PI	0.84 0.84	メタノール	28	15.81
比較例4	150	24	$CH_3C(OCH_3)_3$	49.5	なし		$Mg(OMe)_2$ Bu_4PI	0.84 0.84	メタノール	28	7.56

(注) *1 オルトエステルに対する収率

*2 シス－ジシクロヘキサノ－18－クラウン－6

[0020]Except having made it react for example 972 hours, when reacted completely like Example 1, dimethyl carbonate generated with the yield of 35.9% to the taught alt.methyl acetate.
Except the pressure in the autoclave of example 10 reaction time (150 **) having been 60 atmospheres, when reacted completely like Example 1, dimethyl carbonate generated with the yield of 31.8% to the taught alt.methyl acetate.

[0021]

[Effect of the Invention]According to this invention method, carbon dioxide and carboxylic acid ortho

ester can be made to be able to react, and carbonic ester can be manufactured with high yield. Carbon dioxide does not have toxicity and corrosiveness, it is cheap and this invention method can be enforced industrially and suitably.

[Translation done.]

Exhibit 2

PATENT ABSTRACTS OF JAPAN

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(71)Applicant : SHOWA DENKO KK
(72)Inventor : KOU KO
OGATA FUJIMARO

(54) PRODUCTION OF CARBONIC ACID ESTER

(57)Abstract:
PURPOSE: To efficiently produce a carbonic acid ester from carbon dioxide gas and an alcohol while preventing the deactivation of catalyst by removing produced water according to the progress of the reaction.
CONSTITUTION: A dehydration agent is added to a reactional system and the reaction is carried out while removing water produced according to the progress of the reaction. The dehydrating agent is e.g. orthoformic acid triesters, orthoacetic acid triesters, dicyclohexylcarbodiimide, molecular sieve, cyclopropanone and chloral. This process for the production of a carbonic acid ester is industrially advantageous because the life of the catalyst is remarkably prolonged by the addition of the dehydrating agent compared with the case free from dehydrating agent.

CLAIMS

[Claim(s)]
[Claim 1]A manufacturing method of carbonic ester advancing a reaction removing water by which makes carbon dioxide react to alcohol, faces manufacturing carbonic ester under existence of a catalyst which consists of metallic compounds, makes a dehydrator exist in the system of reaction, and it is generated.
[Claim 2]In the manufacturing method according to claim 1, a dehydrator is a general formula (1).
$R-C(OR^1)_3$ (1)
(among a formula, R expresses hydrogen, an alkyl group, or an aryl group, and R^1 expresses an alkyl group or an aryl group.) -- a manufacturing method being a compound shown.
[Claim 3]A manufacturing method characterized by a dehydrator being a molecular sieve in the manufacturing method according to claim 1.
[Claim 4]In the manufacturing method according to claim 1, a dehydrator is a general formula (2).
$R^2-N=C=N-R^2$ (2)
(R^2 expresses an alkyl group, a cycloalkyl group, or an aryl group among a formula.) -- a manufacturing method being a compound shown.
[Claim 5]A manufacturing method characterized by a dehydrator being cyclopropanone in the manufacturing method according to claim 1.
[Claim 6]A manufacturing method characterized by a dehydrator being trichloroacetic aldehyde in the manufacturing method according to claim 1.

[Translation done.]

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Industrial Application]This invention relates to the method of making carbon dioxide reacting to alcohol and manufacturing carbonic ester. Carbonic ester is useful as raw materials, such as an alkylating agent, a carbonylation agent, a solvent, a gasoline additive, agricultural chemicals, medicine, and polycarbonate manufacture.
[0002]
[Description of the Prior Art]It is known that carbonic ester will be compoundable by various kinds of methods conventionally. For example, the method of making phosgene and alcohol react and compounding carbonic ester with dehydrochlorination is famous. However, by this method, in order to use strong toxic phosgene for a raw material extremely, cautions are required for that handling. The plant-and-equipment investment on security becomes large, and causes a cost hike.
[0003]Carbon monoxide, alcohol, and oxygen are made to react under existence of a metal catalyst as other synthetic methods, and the method of compounding carbonic ester with drying is known. For example, as a method of using a copper compound as a catalyst, the method of making carbon monoxide, alcohol, and oxygen react to JP,45-11129,A under cupric ion existence, such as a cupric chloride and the second copper of bromination, as a catalyst is indicated. The method to which carbon monoxide, alcohol, and oxygen are made to react using the copper salt of the monovalence like bromination, chloridation, or the first copper of perchloric acid as a catalyst is indicated by JP,60-58739,B. Since the activity of carbonic ester generation by these methods is low, there is much amount of the copper compound used, And since it originates in the solubility being small and a catalyst component deposits so much, a device special to reaction operation and recovery recycling of a catalyst is required, There is a fault of that alkyl halide, ester, etc. furthermore carry out many byproductions and carbon monoxide oxidizing by oxygen and generating carbon dioxide. The way a copper system catalyst raises the stability of a catalyst by controlling the moisture in reaction mixture to 3% or less since an activity fall takes place easily with moisture is proposed by JP,1-279859,A.
[0004]The method of making alcohol and carbon dioxide react and on the other hand, compounding carbonic ester is also publicly known. The method of making the bottom of existence of tin ARUKO oxide or titanium ARUKO oxide, alcohol, and carbon dioxide react to JP,56-40707,B, and compounding carbonic ester is indicated. However, by this method, a catalyst is inactivated with the water separated with advance of a reaction, and it does not fully rotate.
[0005]
[Problem(s) to be Solved by the Invention]By the method of making above-mentioned carbon monoxide, alcohol, and oxygen react under existence of a copper system catalyst, and compounding carbonic ester, the carbon monoxide whose toxicity it is comparatively expensive and is strong is used, In order to react under existence of oxygen, there are problems, like that the side reaction by oxidation is hard to be inhibited and poisoning by the water to separate cannot be controlled.
[0006]What comparatively cheap carbon dioxide is used for by the method of compounding carbonic ester for carbon dioxide and alcohol under existence of tin or a titanium system catalyst, That a catalyst is inactivated with the water separated although solved has not still solved some of problems

at the time of compounding carbonic ester from carbon monoxide, alcohol, and oxygen by not making oxygen exist in the system of reaction.
[0007]The purpose of this invention is to provide the method of manufacturing carbonic ester advantageously by facing compounding carbonic ester, using cheap carbon dioxide as a raw material, preventing inactivation of a catalyst, and rotating a catalyst enough.
[0008]
[Means for Solving the Problem]Hereafter, this invention is explained in detail. This invention persons face compounding carbonic ester from carbon dioxide and alcohol, using a metaled alkoxy compound as a catalyst, As a result of examining wholeheartedly how to remove water to separate and prevent inactivation of a catalyst, by removing promptly water which separated a dehydrator by making it exist in the system of reaction, it finds out that catalytic activity is maintained over a long period of time, and came to complete this invention.
[0009]That is, under existence of a catalyst which consists of metallic compounds, this invention makes carbon dioxide react to alcohol, is faced manufacturing carbonic ester, and it relates to a manufacturing method of carbonic ester advancing a reaction, removing water by which makes a dehydrator exist in the system of reaction, and it is generated.
[0010]A catalyst applicable to this invention is a general formula (3).
$(R^3)_m M(OR^4)_{4-m}$ (3)
(R^3 and R^4 show an alkyl group or an aryl group among a formula, m expresses an integer of 0 to 3, and M expresses metal.) -- or general formula (4)
$(R^5)_2MO$ (4)
(R^5 expresses an alkyl group or an aryl group among a formula, and M expresses metal.) -- it is a compound shown. For example, as R^3, R^4, and R^5, methyl, ethyl, n-propyl, i-pull pill, n-butyl, phenyl, tolyl, anisyl, etc. are mentioned. Sn, Ti, etc. are mentioned as M.
[0011]Alcohol of a raw material applicable to this invention has fatty alcohol, especially preferred methanol, although aromatic alcohol, such as unsaturated alcohol, such as fatty alcohol, such as methanol, ethanol, propanol, and butanol, and allyl alcohol, and phenol, is mentioned, for example.
[0012]A dehydrator in this invention is a general formula (1).
$R-C(OR^1)_3$ (1)
(R expresses hydrogen, an alkyl group, or an aryl group among a formula, and R^1 expresses an alkyl group or an aryl group.) -- it is a compound shown, for example, alt.formate, alt.acetate ester, etc. are mentioned.
[0013]Or a dehydrator is a general formula (2).
$R^2-N=C=N-R^2$ (2)
(R^2 expresses an alkyl group, a cycloalkyl group, or an aryl group among a formula.) -- it is a compound shown, for example, dicyclohexylcarbodiimide etc. are raised. Or a molecular sieve, cyclopropanone, or trichloroacetic aldehyde may be sufficient as a dehydrator.
[0014]The number of dehydrators may be one and they may be used combining two or more kinds. a carbon dioxide partial pressure within the system of reaction -- 1 - 250 kg/cm^2 -- it is the range of 10-150kg[/cm] ^{2} preferably. A range of 50-250 ** of reaction temperature is 100-200 ** preferably. If reaction temperature is too low, reaction velocity becomes slow, and a decomposition reaction of carbonic ester generated when too high is promoted, and it is not desirable.
[0015]
[Example]Although an example explains this invention below, this invention is not limited to the range of the following example.
[0016]To the autoclave made from SUS of 100 ml of inner capacity with example 1 agitating device, 10 ml of methanol, The dibutyldimethoxy tin 1.5g was prepared as a catalyst, 8.8 g of alt.formic acid trimethyl was taught as a dehydrator, and after it introduced carbon dioxide from the carbon dioxide cylinder and carbon dioxide replaced the inside of autoclave 3 times, the internal pressure of autoclave was adjusted to 30 kg/cm^2. It heated at 150 **, agitating the inside of autoclave after an

appropriate time, and was made to react for 24 hours. The pressure in the autoclave at the time of a pyrogenetic reaction had reached about 100 kg/cm^2. The carbon dioxide which remains after cooling was emitted and gas chromatography analyzed reaction mixture. The result checked that 5.9 g of dimethyl carbonate generated and methyl formate which is 4.1g was generating. The quantity of the generated dimethyl carbonate was a 13.2 time mol to the added catalyst. That is, it means that the catalyst had rotated 6.6 times.

[0017]To the autoclave made from SUS of 100 ml of inner capacity with example 2 agitating device, 10 ml of methanol, The dibutyldimethoxy tin 1.5g was prepared as a catalyst, 10 g of alt.acetic acid trimethyl was taught as a dehydrator, and after it introduced carbon dioxide from the carbon dioxide cylinder and carbon dioxide replaced the inside of autoclave 3 times, the internal pressure of autoclave was adjusted to 30 kg/cm^2. It heated at 150 **, agitating the inside of autoclave after an appropriate time, and was made to react for 8 hours. The pressure in the autoclave at the time of a pyrogenetic reaction had reached about 100 kg/cm^2. The carbon dioxide which remains after cooling was emitted and gas chromatography analyzed reaction mixture. The result checked that 2.2 g of dimethyl carbonate generated and methyl acetate which is 1.9g was generating. The quantity of the generated dimethyl carbonate was a 5.0 time mol to the added catalyst. That is, it means that the catalyst had rotated 2.5 times.

[0018]To the autoclave made from SUS of 100 ml of inner capacity with example 3 agitating device, 10 ml of methanol, The dibutyldimethoxy tin 1.5g was prepared as a catalyst, DCC (dicyclohexylcarbodiimide)8.5g was taught as a dehydrator, and after it introduced carbon dioxide from the carbon dioxide cylinder and carbon dioxide replaced the inside of autoclave 3 times, the internal pressure of autoclave was adjusted to 30 kg/cm^2. It heated at 150 **, agitating the inside of autoclave after an appropriate time, and was made to react for 8 hours. The pressure in the autoclave at the time of a pyrogenetic reaction had reached about 100 kg/cm^2. The carbon dioxide which remains after cooling was emitted and gas chromatography analyzed reaction mixture. As a result, 2.1 g of dimethyl carbonate generated. The quantity of the generated dimethyl carbonate was a 4.6 time mol to the added catalyst. That is, it means that the catalyst had rotated 2.3 times. [0019]To the autoclave made from SUS of 100 ml of inner capacity with example 4 agitating device, 23 ml of butanol, The dibutyldimethoxy tin 1.5g was prepared as a catalyst, DCC(dicyclohexylcarbodiimide)8.5g was taught as a dehydrator, and after it introduced carbon dioxide from the carbon dioxide cylinder and carbon dioxide replaced the inside of autoclave 3 times, the internal pressure of autoclave was adjusted to 30 kg/cm^2. It heated at 150 **, agitating the inside of autoclave after an appropriate time, and was made to react for 8 hours. The pressure in the autoclave at the time of a pyrogenetic reaction had reached about 100 kg/cm^2. The carbon dioxide which remains after cooling was emitted and gas chromatography analyzed reaction mixture. As a result, 4.1 g of dibutyl carbonate generated. The quantity of the generated dibutyl carbonate was a 4.8 time mol to the added catalyst. That is, it means that the catalyst had rotated 2.4 times.

[0020]To the autoclave made from SUS of 100 ml of inner capacity with a comparative example agitating device, 10 ml of methanol, The dibutyldimethoxy tin 1.5g was prepared as a catalyst, and after it introduced carbon dioxide from the carbon dioxide cylinder and carbon dioxide replaced the inside of autoclave 3 times, the internal pressure of autoclave was adjusted to 30kg[/cm] ^{2}. It heated at 150 **, agitating the inside of autoclave after an appropriate time, and was made to react for 24 hours. The pressure in the autoclave at the time of a pyrogenetic reaction had reached 100 kg/cm^2. The carbon dioxide which remains after cooling was emitted and gas chromatography analyzed reaction mixture. The result checked that 0.4 g of dimethyl carbonate was generating. The quantity of the generated dimethyl carbonate was a 0.8 time mol to the added catalyst. That is, it means that the catalyst had rotated 0.4 times.

[0021]

[Effect of the Invention]As explained above, the result whose number of rotations of a catalyst improves remarkably by addition of the dehydrator to the system of reaction is obtained, the life of a catalyst is improved remarkably, and the manufacturing method of carbonic ester excellent in the industrial target can be provided.

[Translation done.]

Exhibit 3

DECLARATION OF TRANSLATOR

I, Osamu MAEDA, c/o the Inoue & Associates of 3rd Floor, Akasaka Habitation Building, 3-5, Akasaka 1-chome, Minato-ku, Tokyo, Japan do solemnly and sincerely declare that I am conversant with the Japanese and English languages and that the attached text is a partial English translation of Unexamined Japanese Patent Application Laid-Open Specification No. Sho 54-3012 (JP 54-3012), and believe that the translation is true and correct.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

February 20, 2008
(Date)

Osamu Maeda
Osamu MAEDA

Partial English Translation of Unexamined Japanese Patent Application Laid-Open Specification No. Sho 54-3012 (JP 54-3012)

(I) At page 1, left upper column, lines 1-3

(54) Method for producing a carbonate

(21) Application number: Sho 52-68310

(22) Date of filing: June 9, 1977

(II) Page 1, Scope of claims for patent

1. A method for producing a carbonate, comprising reacting a hydroxy compound with carbon dioxide in the presence of a tin alkoxide or a tetraalkoxytitanium,

wherein said tin alkoxide is represented by the following formula (I):

$$R_{3-\ell}Sn(OR^1)_{1+\ell} \qquad (I)$$

wherein each R represents a hydrocarbon group, each R^1 represents a hydrocarbon group, ℓ represents an integer of from 0 to 3, and two R^1 groups may be the same alkylene group, and

wherein said tetraalkoxytitanium is represented by the following formula (II):

$$Ti(OR^2)_4 \qquad (II)$$

wherein each R^2 represents a hydrocarbon group.

2. The method according to claim 1, wherein said hydroxy compound is represented by the following formula (III):

$$R^3(OH)_m \qquad (III)$$

wherein R^3 represents a hydrocarbon group having m value, and m represents an integer of 1 or more.

(III) Page 3, left upper column, lines 1-5

In the method of the present invention, a hydroxy compound is reacted with carbon dioxide. The term "hydroxy compound" means an organic compound having a hydroxyl group bonded to a carbon atom. Examples of hydroxy compounds include alcohols and phenolic compounds.